MECHANICAL RESONANCE DISPERSION AND STRESS-STRAIN BEHAVIOR OF SEVERAL IONIC SINGLE CRYSTALS

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SUMMARY

The tangent modulus of the compressive stress-strain curve, at any stress level up to the inflection point, agrees well with the low-frequency dynamic modulus determined at the corresponding stress level by the Fitzgerald method. This agreement is attributed to the fact that in this stress range both the "static" and dynamic tests sample the internal dislocation configuration that gives rise to the foot on the stress-strain curve.

Above the inflection point the dynamic modulus is higher than the tangent modulus determined at the same stress level. Here the lack of agreement is due to a difference in the tests at these stress levels. The dynamic test still samples the dislocation configuration, but the static tests reflect the effects of gross plastic-deformation processes.

For the first compression unloading curve and successive loadings and unloadings, the dynamic modulus is lower than the tangent modulus determined at corresponding stresses. These deviations are the result of the dynamic nature of the Fitzgerald measurements.

Mechanical resonance dispersions observed in sodium chloride, potassium chloride, potassium bromide, and lithium fluoride are qualitatively similar to those reported by Fitzgerald for other single crystals. The frequency spectrum of J' (storage compliance) and J'' (loss compliance), that is, the resonance structure, depends on static compressive-clamping stress. Furthermore, the effects of increasing the static stress are not reversible upon decreasing the static stress.

INTRODUCTION

Recent measurements (refs. 1 and 2) have established that S-shaped compressive stress-strain curves are characteristic of single-crystal sodium chloride, potassium bromide, lithium fluoride, potassium chloride, magnesium oxide, and copper. The initial concave-upward portion of the curve, termed the foot, was shown to be a manifestation of a real deformation mode of the material under test. Furthermore, the magnitude of the foot was shown to vary

with testing rate, specimen pretreatment, and prestrain. A qualitative explanation of these results was presented in terms of a model involving internal stresses and dislocation velocities. While the proposed model appeared to be consistent with most dislocation concepts, more information is needed before a detailed description can be obtained and the model can be tested.

Additional information regarding the mechanisms that account for the foot of the stress-strain curve should be obtainable from a suitable dynamic test. The determination of dynamic properties as functions of position on the stress-strain curve would be most desirable. Several features of the work of Fitzgerald (refs. 3 and 4) appear interesting in this regard. First, the dynamic shear apparatus developed by Fitzgerald (refs. 5 and 6) subjects a sample, clamped statically by a compressive stress, to a sinusoidal shear stress in the frequency range nominally from 50 to 5000 cps. With this apparatus the components of the complex dynamic shear compliance $J^* = J' - iJ''$ can be determined, where J' is the storage compliance and J'' is the loss compliance. Thus, by varying the clamping stress, dynamic properties can be measured as functions of position on the stress-strain curve.

A second interesting aspect of the work of Fitzgerald is the low-frequency (≈ 400 cps) dynamic moduli reported for several metal and ionic crystals (refs. 3 and 4). These moduli are much lower than expected on the basis of elastic-constant calculations (ref. 7), and for sodium chloride, a Fitzgerald dynamic modulus is in fair agreement with the tangent modulus of the stress-strain curve determined at corresponding stress levels in the foot region (ref. 8).

Finally, the frequency spectra of J' and J'' reported by Fitzgerald (refs. 3 and 4) display multiple dispersions of the resonance type. Values of the storage compliance J' rise to a maximum and then drop, passing through a minimum as a narrow dispersion region is traversed in the direction of increasing frequency. The nonresonance value of J' is lower on the high-frequency side. Such dispersions could account for the low dynamic moduli measured at low frequencies.

Based on these considerations, a comparison of Fitzgerald-type dynamic measurements and compressive stress-strain tests was undertaken for a group of ionic single crystals. The low-frequency dynamic moduli were measured at various static clamping stresses and compared with the tangent moduli of the compressive stress-strain curve measured at corresponding stresses. Also, the components of the complex shear compliance were examined as functions of frequency and applied static clamping stress.

EXPERIMENTAL

All crystals used in this investigation were purchased from the Harshaw Chemical Company in the form of cleaved rectangular prisms. The pair of specimens required for the dynamic measurements was cleaved in this laboratory from the same parent prism. Specimen height was approximately 5 millimeters, and the cross section varied from 6.5 by 6.5 to 6.5 by 19.0 millimeters.

The specimens for the compressive stress-strain tests were cleaved to a cross section measuring approximately 6.5 by 6.5 millimeters and to a height of 19.0 millimeters. To assure flat and parallel end faces, the specimens were polished according to the procedure described in reference 1.

All the materials investigated were studied in the as-cleaved, annealed, and X-irradiated states. The annealing procedure consisted of heating the specimen to the annealing temperature of 100° C below the melting point (at a maximum rate of 50° C/hr), holding it at the annealing temperature for 12 hours, and then cooling it to room temperature at a maximum rate of 45° C per hour. All anneals were performed in a tube furnace that was open to air.

X-ray doses were obtained with the specimen rotating in the beam emanating from a tungsten-target, beryllium-window, X-ray tube operated at 250 kilovolts and 8 milliamperes. The specimen-to-window distance was 12 inches giving a dose rate of 2×10^3 roentgens per minute. All irradiations were done in air and in the dark.

TEST METHOD

The complex dynamic shear compliance $J^* = J' - iJ''$ consists of two components: J' is the component of shear strain in phase with the shear stress divided by the stress, and J'' is the component of the shear strain 90° out of phase with the shear stress divided by the stress. The elastic energy stored and recovered during each cycle of deformation is proportional to J', and hence J' is called the storage compliance; J'' is proportional to the energy dissipated during each cycle and is called the loss compliance. Similarly, if the strain is taken as a reference and the stress is considered to consist of in-phase and quadrature components, a complex dynamic shear modulus $G^* = G' + iG'' = 1/J^*$ results, where G' is the dynamic modulus and G'' is the loss modulus.

The components of J^* were measured by the electromagnetic transducer method of Fitzgerald and Ferry (ref. 5). The apparatus employed was identical to that of Fitzgerald. Basically, the transducer consists of an aluminum driving tube to which are attached two circumferential coils. This driving tube is suspended by wires in a radial magnetic field. An inertial mass is suspended concentrically with the driving tube and is so arranged that the specimens to be tested are compressed statically between the driving tube and the mass by a wedge and screw arrangement. When a sinusoidal current is passed through one of the coils on the driving tube, the samples are sheared sinusoidally in a direction perpendicular to the static compressive-clamping stress. The second coil on the driving tube acts as a pickup coil; this coil is one arm of an alternating-current bridge and its total dynamic impedance is measured. The mechanical impedance of the sample is then determined by the vectorial subtraction of the impedance of the system from the total measured impedance as outlined by Fitzgerald (ref. 5).

All dynamic measurements reported here were made at room temperature and at a constant peak driving force of 3.5×10^3 dynes. Resonance peaks were approximately located by observing on an oscilloscope the Lissajous figure

produced when the driving current (proportional to driving force) and pickup coil voltage (proportional to velocity) were applied to the scope deflection plates. Although the transducer was not designed to give quantitative data above 5×10^3 cps, resonances up to 10^4 cps could be observed with the oscilloscope.

The static compressive-clamping stress was measured by means of strain gages bonded to the driving tube. The load-strain curve for the driving tube was determined by deadweight loading prior to installation of the driving tube in the transducer. The load-strain curve was linear to within ±3 percent.

The accuracy of the dynamic measurements was estimated to be ±5 percent. The accuracy of the static clamping stress determination was limited by strain measurements and the determination of the slope of the driving-tube load-strain curve. Accuracy was estimated to be ±0.5 grams per square millimeter.

The dynamic modulus G' was determined as a function of static compressive-clamping stress σ_c; σ_c was varied in steps from a few grams per square millimeter to beyond the macroyield region of the stress-strain curve. To enable the measurement of the small static stresses in the transducer, relatively large cross-sectional area specimens were employed; this resulted in a loss of accuracy in the dynamic measurements below about 500 cps. Since no resonances were observed on the oscilloscope below 500 cps, this was accepted as the low-frequency limit for the dynamic measurements.

For the comparison of the dynamic modulus with the slope of the compressive stress-strain curve, G' was computed from the average compliance levels of the frequency spectra in the flat region between 500 cps and the first resonance. To compare the dynamic measurements with the stress-strain curve, G' was converted to a dynamic Young's modulus E' by using $E' = 2G'(1 + \mu)$, where μ is Poisson's ratio.
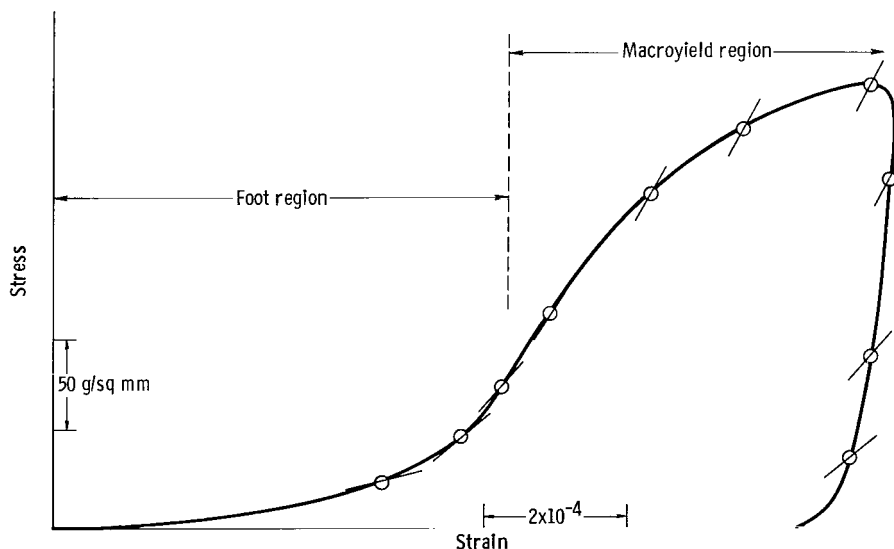

Figure 1. - Annealed lithium fluoride.

Because the initial slope of the compressive stress-strain curve is loading-rate dependent (ref. 1), the rates for the compression tests were adjusted to correspond to the dynamic rates.

RESULTS AND DISCUSSION

Dynamic Modulus

A comparison of the measured dynamic modulus with the slope of the compressive stress-strain curve is shown in figure 1 for annealed lithium fluoride. From this figure it is evident that the dynamic modulus E' agrees with the tangent modulus for stresses up to the inflection point of the stress-strain curve. Above the inflection point and through the onset of macroyielding, the dynamic moduli are seen to take on an almost constant value that is greater than the corresponding value of the tangent moduli. For the unloading curve the dynamic moduli are lower than the corresponding tangent values. Similar comparisons made for as-cleaved and X-irradiated as well as annealed sodium chloride, potassium bromide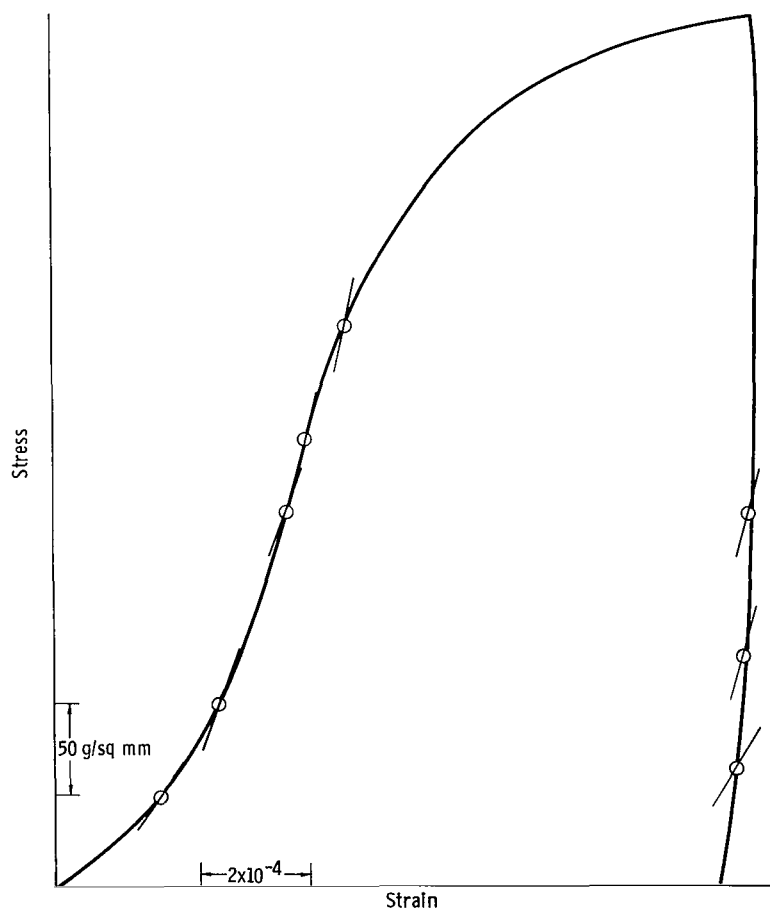, lithium fluoride, and potassium chloride showed the same general results. Data for irradiated and as-cleaved sodium chloride are shown in figures 2 and 3(a), respectively. In figure 2 it is seen that the dynamic moduli again agree with the tangent moduli up to the inflection point, which is now a factor of five higher than for as-cleaved sodium chloride. In figure 3(a) the initial part of the stress-strain curve is shown on an expanded scale, and the agreement between E' and the corresponding tangent modulus is seen to be excellent for the loading curve. (In this case the specimens were not loaded into the macroyield region).

The comparison of dynamic with tangent moduli was also made on specimens subjected to successive loading-unloading cycles. Again the specimens were not loaded into the macroyield region. A pair of typical results is shown in figures 3(b) and (c) for as-cleaved potassium

Figure 2. – X-irradiated sodium chloride. Dose, 24x10^5 roentgens.

(a) Sodium chloride.

(b) Potassium bromide.
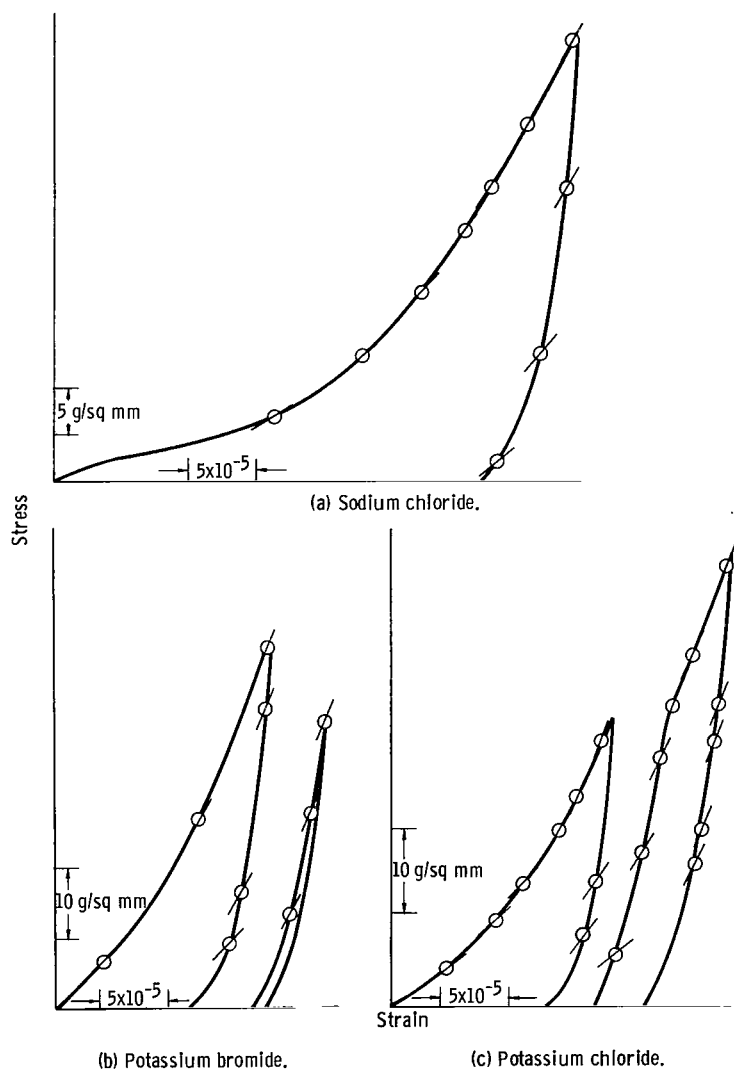
(c) Potassium chloride.

Figure 3. - Stress-strain curves for as-cleaved compounds. Slopes determined by Fitzgerald-type measurements at approximately 500 cps.

bromide and potassium chloride, respectively. In both of these figures it is apparent that the dynamic moduli agree fairly well with the tangent moduli for the first loading. During the first unloading, the dynamic moduli are seen to be lower than the tangent moduli. For the second loading the dynamic moduli are lower than the tangent moduli for stresses below the first-loading maximum. Above the first-loading maximum stress (but still below the inflection point), the dynamic moduli again agree with the tangent moduli (fig. 3(c)).

The deviation between dynamic and tangent moduli observed above the inflection point is believed to be due to an intrinsic difference between the two tests. In the compression test gross plastic-flow processes predominate beyond the inflection point. The dynamic test, by virtue of the very small amplitude of the shear stress and strain involved, only detects the equilibrium (at such static stress levels) dislocation configuration and not processes such as multiplication.

On initial loading below the inflection point, the dynamic moduli are found to correlate with the tangent moduli. In this stress range the two tests are essentially the same; that is, they are responding to the same processes. The shape of the stress-strain curve below the inflection point is primarily determined by the way the dislocation configuration changes with stress. In this region both moduli are a measure of the configuration.

As shown in figures 3(b) and (c) the dynamic moduli do not agree with the tangent moduli on the first unloading curve and on subsequent loading and unloading curves. These deviations can be explained hypothetically in the following manner. In the static test, the dislocation configuration is determined by the maximum stress applied; that is, only slight relaxation occurs
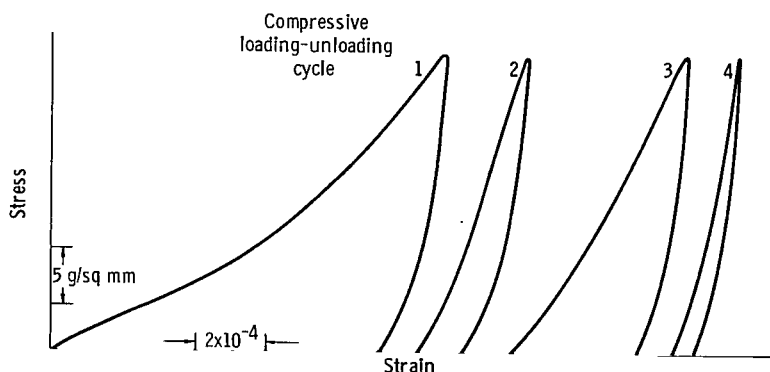
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Figure 4. - Successive loading-unloading cycles for as-cleaved sodium chloride showing effects of dynamic shaking.

upon stress removal. In the dynamic test, the repetitive stress reversal causes the dislocation network to relax into a new configuration. This configuration is determined by the static stress level about which the dynamic measurements are made. Data (fig. 4) from an experiment performed by the authors of this report support this hypothesis.

Two cycles of compressive loading-unloading were executed in the tensile test apparatus. These two cycles are shown in figure 4 as cycles 1 and 2. Cycle 2 is narrower or more closed than cycle 1, and the residual strain associated with cycle 2 is considerably less than that associated with cycle 1. As shown in reference 1, succeeding cycles to or below the maximum stress of cycle 1 would close even more, and eventually a closed loop with no residual strain would result. After cycles 1 and 2 were performed, the specimen was transferred to the Fitzgerald apparatus. It was clamped to a stress level about 10 percent lower than the maximum stress of cycle 1. The specimen was then sheared dynamically for several minutes at various frequencies from 500 to 5000 cps. Next, the clamping stress was reduced to about one eighth the maximum stress of cycle 1, and again the specimen was sheared dynamically for several minutes. This shaken specimen was then returned to the tensile test apparatus, and two more compressive loading-unloading cycles were performed (cycles 3 and 4 in fig. 4). It is immediately evident from figure 4 that the tangent moduli for cycle 3 are lower than those for cycle 2 at corresponding stress levels. These results are in general agreement with the cyclic softening reported in other investigations (refs. 1, 2, and 9) and tend to support the explanation given previously for the lack of agreement between dynamic and tangent moduli for first-compression unloading and successive loadings and unloadings.

Frequency Spectra of J' and J''

Measurements of the complex shear compliance at closely spaced frequencies in the range from 500 to 5000 cps have revealed that single-crystal sodium chloride, potassium bromide, potassium chloride, and lithium fluoride exhibit multiple-frequency dispersions of the resonance type. The details of the frequency spectra, namely, the number, location and magnitude of resonances, were found to depend on the static clamping stress level at which the measurements were made.

The frequency spectra of the components of the complex shear compliance are shown in figure 5 for an annealed potassium chloride single crystal. The static compressive-clamping stress σ_c was approximately 40 grams per square millimeter. Four prominent resonances are evident at about 1525, 2750, 3300, and 3425 cps. Associated with each resonance dispersion is a change in the
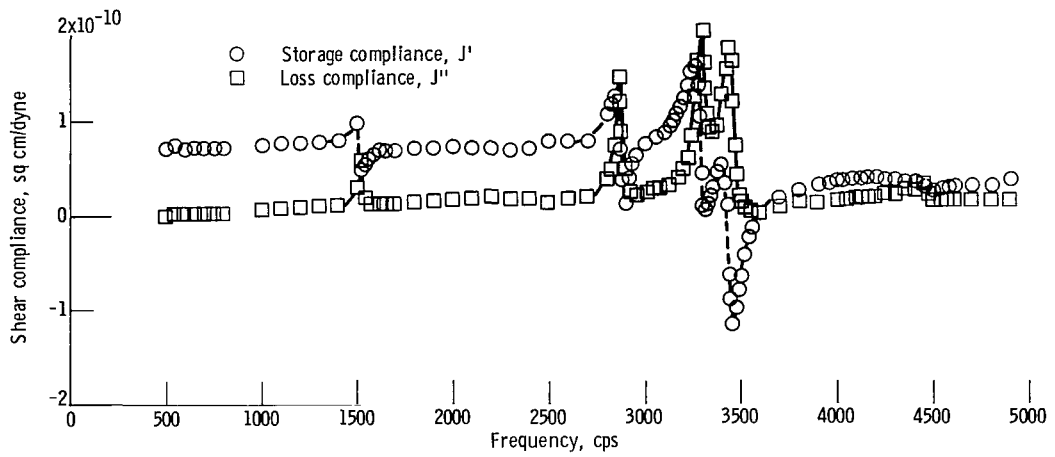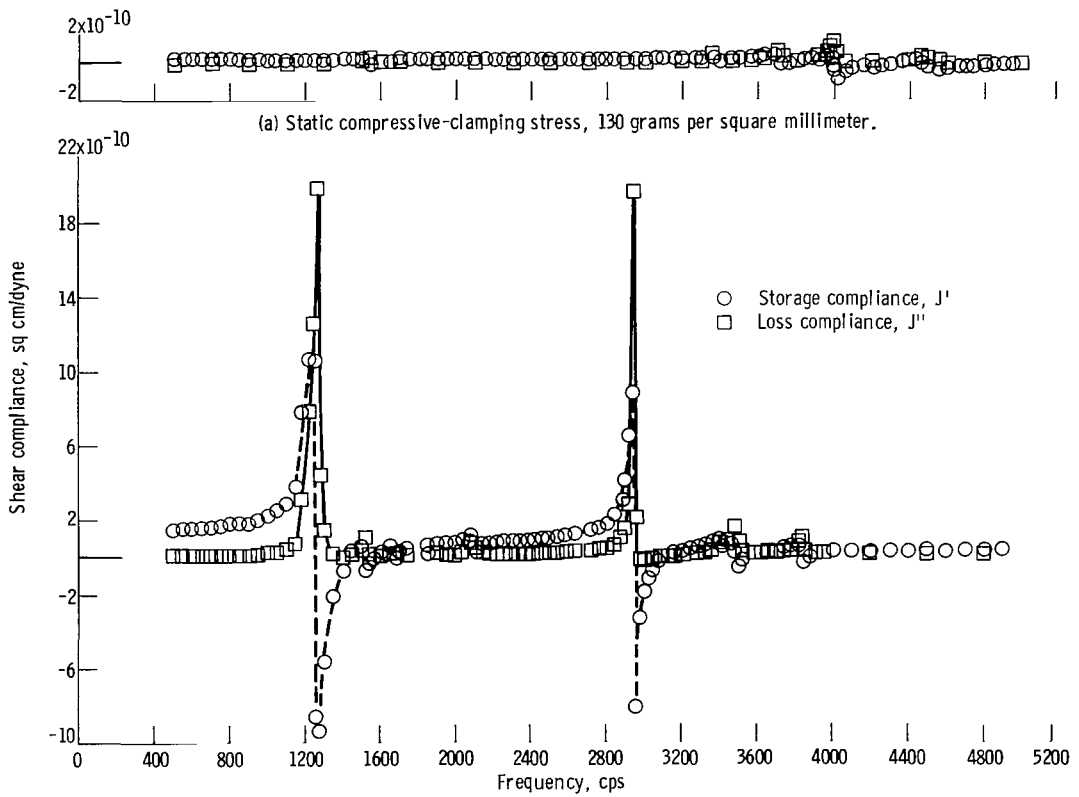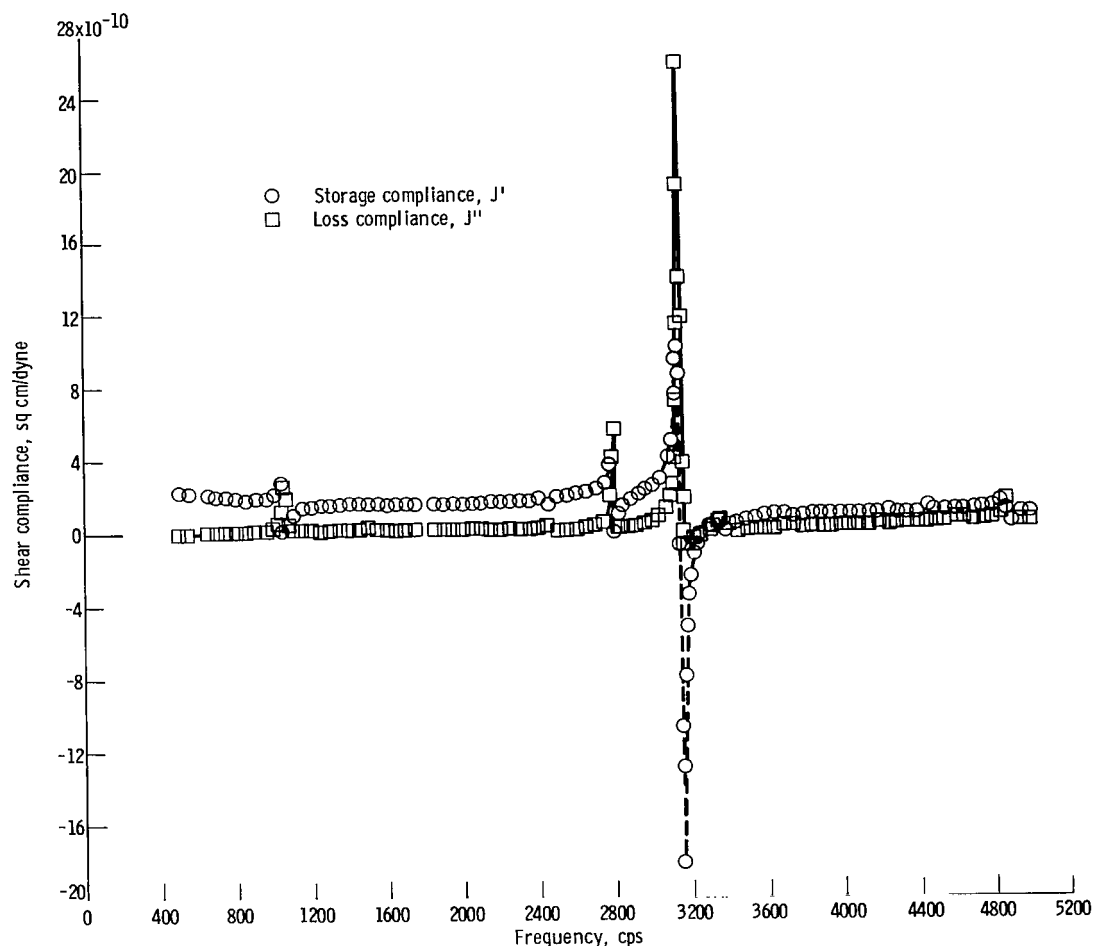
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Figure 5. - Annealed potassium chloride. Static compressive-clamping stress, 40.5 grams per square millimeter.

(a) Static compressive-clamping stress, 130 grams per square millimeter.

(b) Static compressive-clamping stress, 28.9 grams per square millimeter.
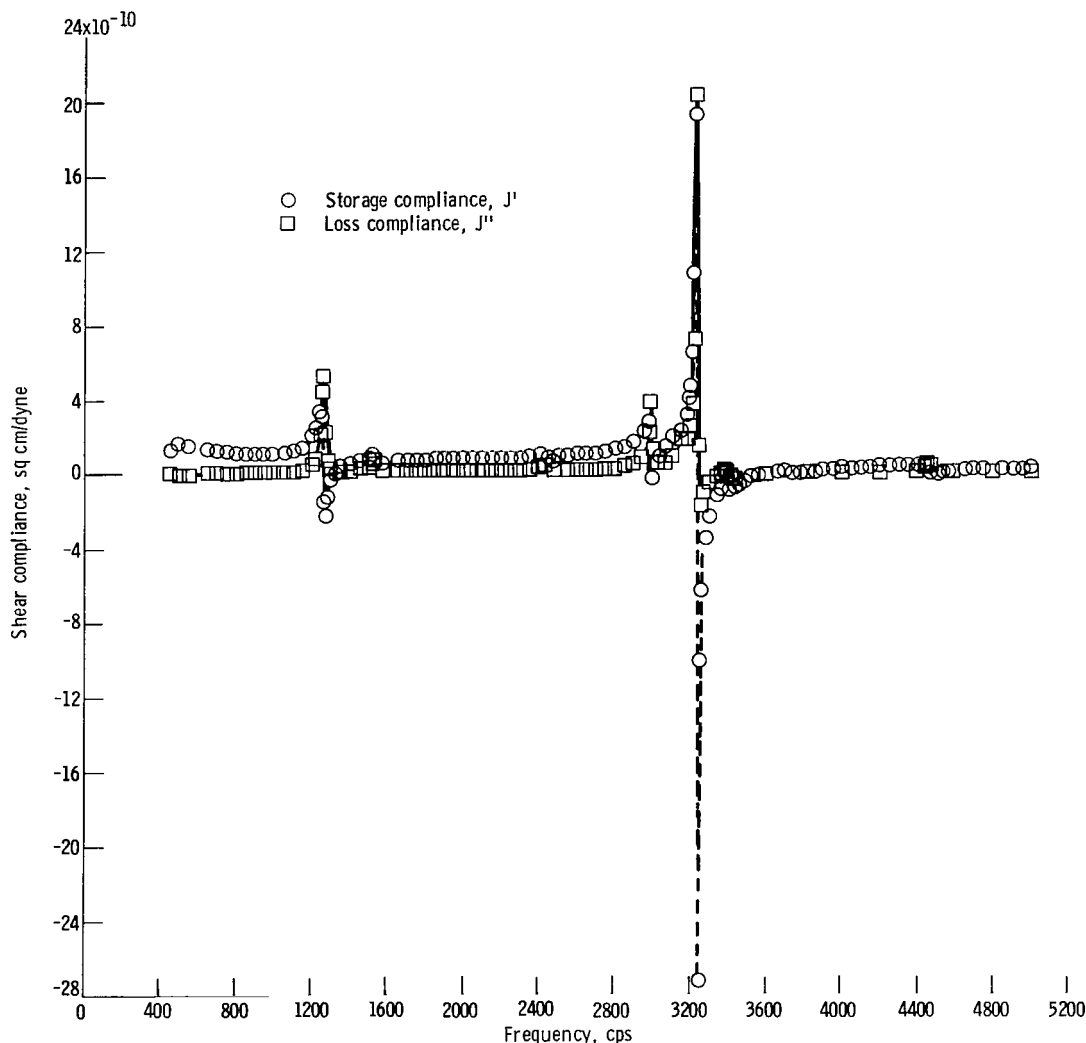
Figure 6. - Annealed lithium fluoride.

(a) Static compressive-clamping stress, 8.9 grams per square millimeter.

Figure 7. – As-cleaved potassium bromide.

general level of J'. For example, at 2000 cps, J' is 0.7×10^{-10} square centimeter per dyne, but at 4100 cps on the other side of the multiple resonances, J' is 0.4×10^{-10} square centimeter per dyne. The amount of change in J' level from one side of a resonance to the other side depends on the magnitude of the dispersion, and in many instances J' was found to change by as much as a factor of five.

The changes produced in resonance structure by varying σ_c can be summarized as follows: (1) With an increase in σ_c, some resonances move up in frequency, and some peaks either vanish or move beyond the high-frequency limit of investigation (5000 cps). (2) Often new peaks appear or at least become distinguishable with increasing σ_c. (3) Some peaks increase in height with increasing σ_c. (4) Changes in resonance structure are not completely reversible with increasing and decreasing stress. (5) The nonresonance value of J'' decreases with increased clamping stress. (6) Often negative values of J'' on the high-frequency side of major resonances are found to develop with increasing σ_c. Examples of these findings can be observed in the spectra shown in figures 6 to 9.

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(b) Static compressive-clamping stress, 31.3 grams per square millimeter.

Figure 7. - Concluded. As-cleaved potassium bromide.

Figure 6 shows the frequency spectra of J' and J'' at two stress levels for annealed lithium fluoride. At the low stress level (below inflection point of compressive stress-strain curve) two large peaks (at approximately 1270 and 2950 cps) and five small peaks are evident. At the high-stress level, however, no large peaks are observed, and five very small resonances can be seen at 1500, 3400, 3700, 4000, and 4500 cps. At an intermediate stress level (57.7 g/sq mm) it was noted that the resonance at 1270 cps disappeared and the resonance at 2950 cps decreased in magnitude by a factor of 10 and increased in frequency. This resonance appears at a stress level of 130 grams per square millimeter at 3400 cps.

When the clamping stress was increased from 8.9 to 31.3 grams per square millimeter, the resonance peaks shown in figure 7 for as-cleaved potassium bromide were seen to move up in frequency, and the resonance at 1100 cps
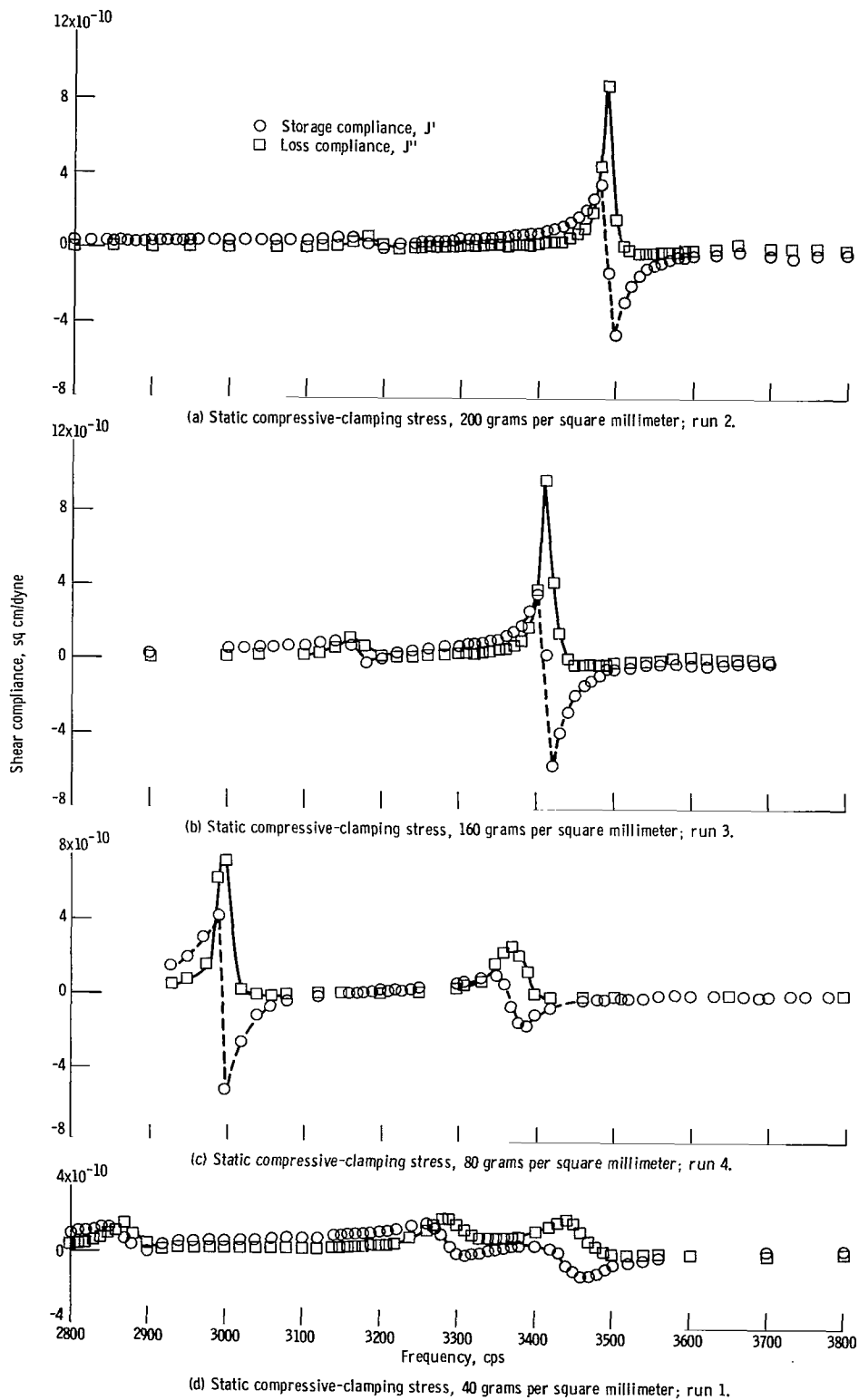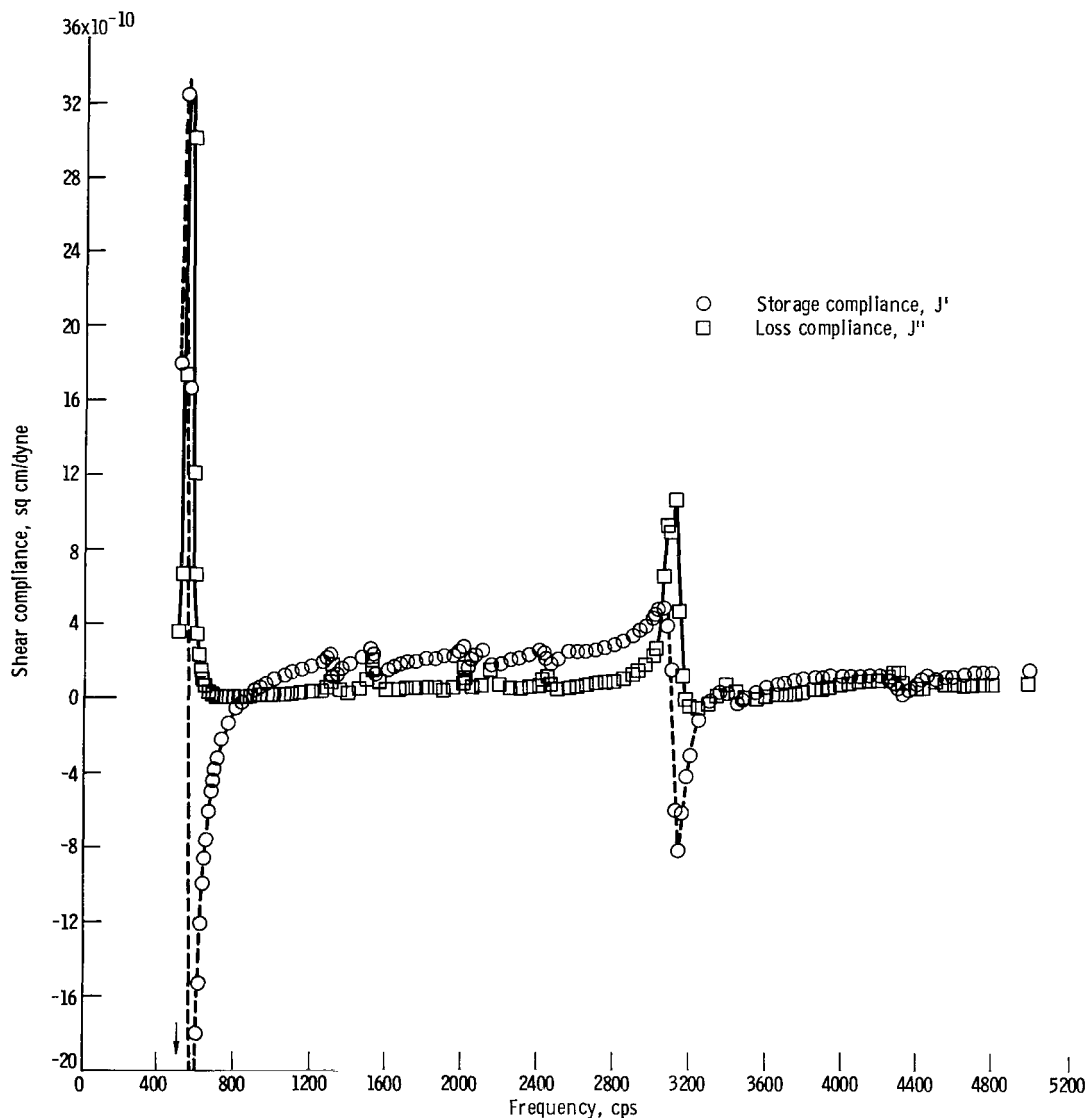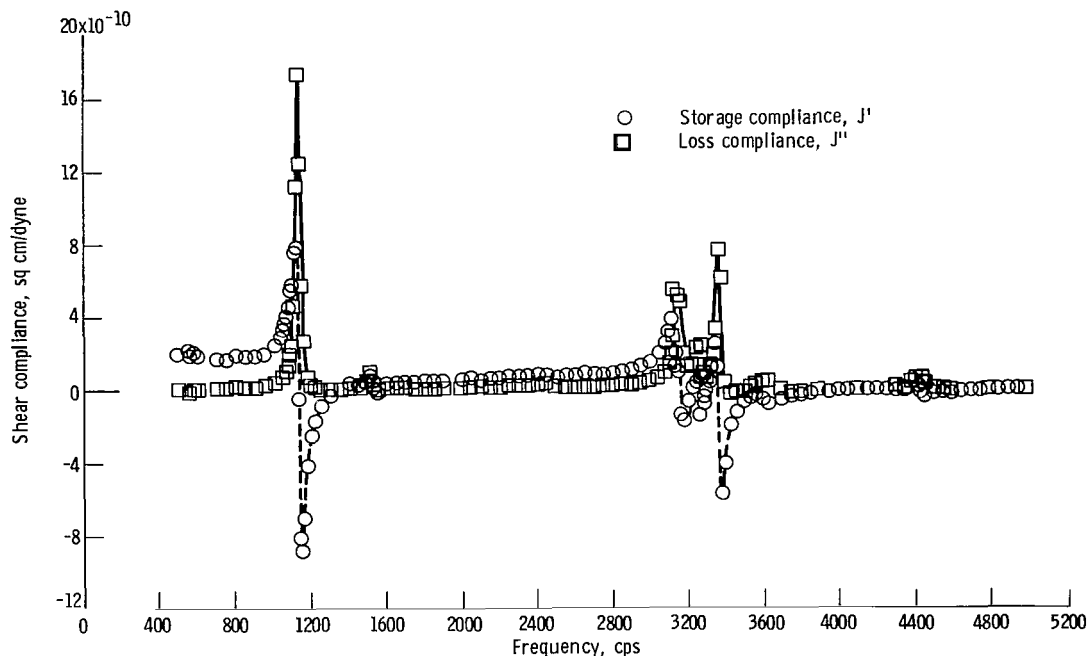
(a) Static compressive-clamping stress, 200 grams per square millimeter; run 2.

(b) Static compressive-clamping stress, 160 grams per square millimeter; run 3.

(c) Static compressive-clamping stress, 80 grams per square millimeter; run 4.

(d) Static compressive-clamping stress, 40 grams per square millimeter; run 1.

Figure 8. - Annealed potassium chloride.

(a) Static compressive-clamping stress, 6.8 grams per square millimeter.

Figure 9. – As-cleaved sodium chloride.

increased in height. Also, the magnitude of the emission (negative J'') observed at the high-frequency side of the peak near 3200 cps was seen to increase by more than a factor of two for the increase in static clamping stress.

The effects of increasing and decreasing the clamping stress are shown in figure 8 for a group of resonances in annealed potassium chloride. In this figure it can be seen that the changes produced by increasing σ_c are not reversible upon decreasing σ_c; that is, the resonances at 3300 to 3500 cps increase with increasing stress, while the resonances at 2900 to 3200 cps increase with decreasing stress.

The general features of the frequency spectra of J' and J'' observed

(b) Static compressive-clamping stress, 27.3 grams per square millimeter.

Figure 9. – Concluded. As-cleaved sodium chloride.

here for several ionic single crystals are the same as those reported by Fitzgerald for other materials (refs. 3 and 4). Considerable controversy exists regarding the origin of the resonances observed with the Fitzgerald test method. Considering the changes produced in the resonance structure by increasing and decreasing the stress together with the complete disappearance of the large resonance in lithium fluoride when σ_c was increased from about 30 to 130 grams per square millimeter, the authors conclude that the resonances are characteristic of intrinsic material properties and not artifacts of the Fitzgerald test as suggested by other experiments (refs. 10 to 12).

The origin of the observed resonances probably lies in the internal dislocation arrays. The configuration of these arrays is known to be altered by stress; thus, one would expect the resonance structure to change with static stress because the dynamic test samples the internal configuration existing at the stress level at which the test is made. The fact that multiple resonances are observed indicates that different configurations respond at different velocities. At low testing frequencies the imposed velocity of deformation is low enough that all segments can respond. As the velocity (or frequency) is increased, preferred velocities are reached; hence, resonances are observed. At frequencies above each resonance the velocity is too high for that segment to respond, and a decrease in J' results. This decrease in J' means that G' has increased; that is, the material behaves a little more elastically because one more nonelastic component cannot respond.

In addition to the changes in resonance structure produced by varying σ_c noted previously, changes associated with the geometric orientation of the specimen were observed. One of the spectra for sodium chloride shown in

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figure 9 has a large resonance at about 600 cps. For this spectrum the clamping stress was 6.8 grams per square millimeter. When the stress was increased to 27.3 grams per square millimeter, this resonance moved up to about 1100 cps. This specimen had a 0.25X0.75-inch cross section and was sheared dynamically in a direction normal to the 0.75-inch dimension. When the specimen was rotated 90° and sheared dynamically parallel to the 0.75-inch dimension, the resonance in the 600 to 1100 cps (depending on stress) completely disappeared. Similar geometric orientation dependencies (involving other resonance frequencies) have been observed in potassium chloride and potassium bromide. While the exact reason for the effect is not completely understood at the present time, it is believed that interactions of resolved dynamic-shear slip systems and preferred compressive slip systems are involved. More intensive investigations into this effect are currently in progress.

CONCLUSIONS

The low moduli measured by the Fitzgerald method appear to be completely compatible with the measured stress-strain behavior. The low moduli are probably the result of many mechanisms involving plastic deformation, each being manifested by an observed resonance-type dispersion. As reported herein, the dynamic modulus increases each time a resonance is passed through, and this means that the particular plastic mechanism responsible for that resonance cannot respond at higher frequencies. On this basis it is not unreasonable to expect that if the complete spectrum to ultrasonic frequencies were available, the modulus would be seen to step up to the usually observed ultrasonic value.

The authors feel that the results presented herein substantiate the validity of the Fitzgerald-type measurements and that this method should provide a valuable tool in studying deformation and defining processes involved.

Lewis Research Center
    National Aeronautics and Space Administration
        Cleveland, Ohio, September 30, 1964

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